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Production of ^{51}Cr neutrino and ^{144}Ce antineutrino generators for SoX and CeLAND experiments

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Search for a hypothetical (sterile?) forth ν state with $\Delta m^2 \approx 0,5 – 3,0\ eV^2$

Two approaches:

– ***using*** $\boldsymbol{\nu_e}$

- detection by elastic scattering of $\boldsymbol{\nu_e}$ on electrons :
  Borexino, KamLAND, others LS detectors) **or**
- detection through CC : $\nu_e + {}^{Z}N_{N\text{-}Z} \rightarrow {}^{Z+1}N_{N\text{-}Z} + e^-$:
  SAGE and LENS detector):

Need strong 5÷10 MCi $\boldsymbol{\nu_e}$ ***sources***;

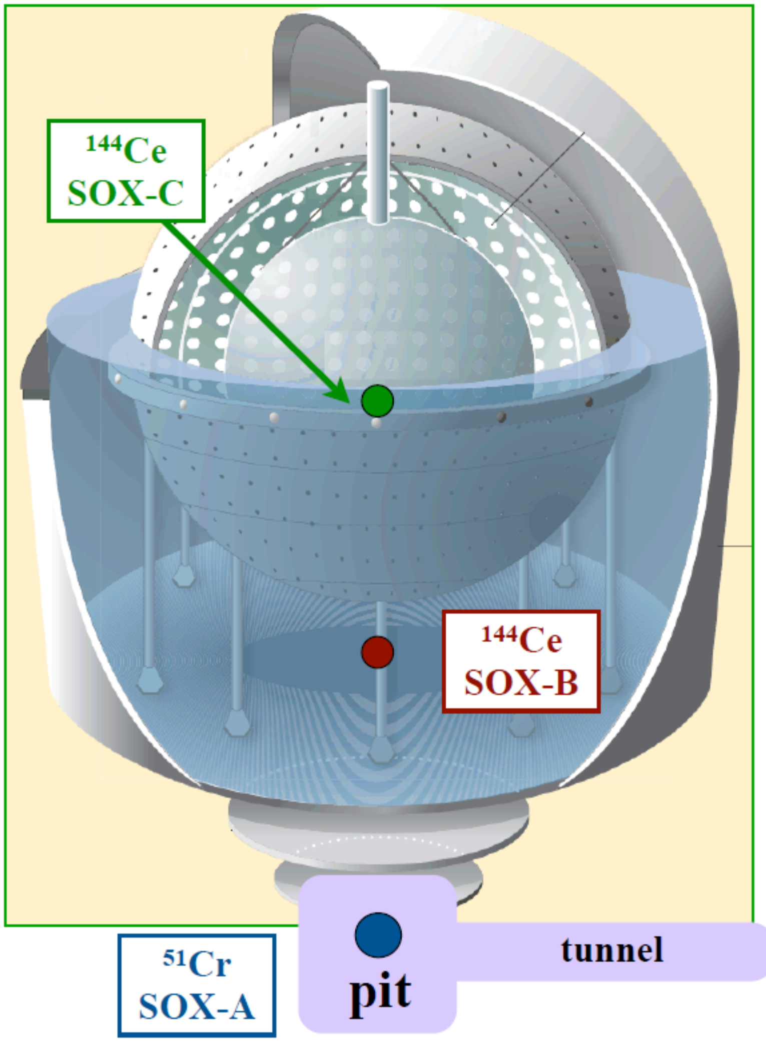
- using ***anti***$\boldsymbol{\nu_e}$ IBD on free protons :

Borexino, KamLAND, LS detector and LENS

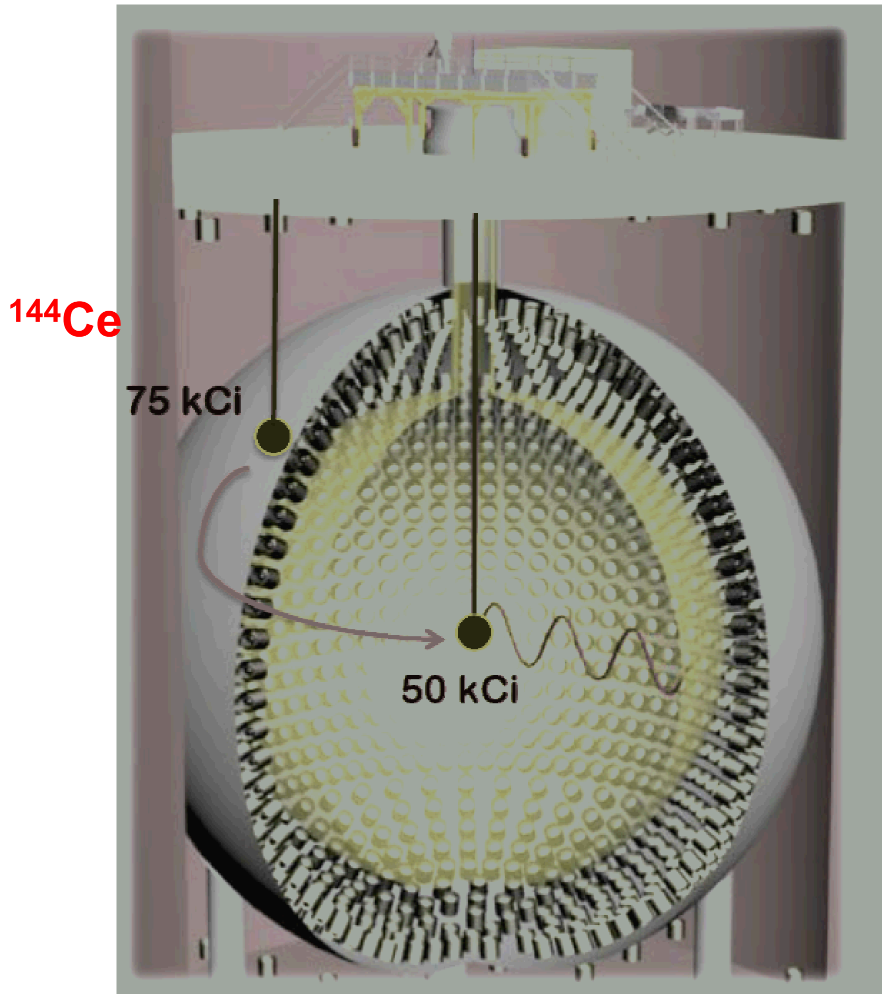
Need ≈ 100 kCi $\boldsymbol{\bar{\nu}_e}$ ***sources***

Call for ^{51}Cr and ^{144}Ce (anti)neutrino sources

SOX: Three Phases



CeLAND: Two phases

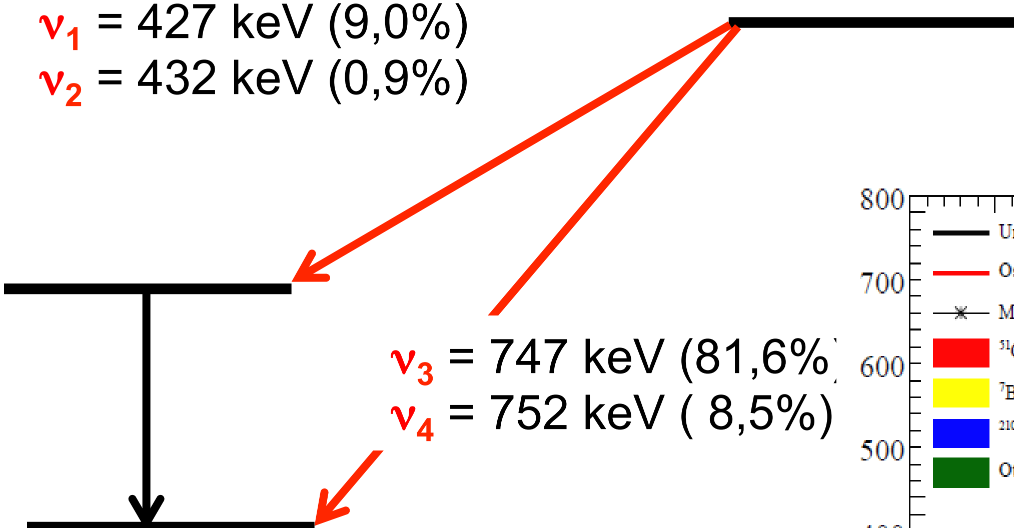


Recipe for a neutrino generator ^{51}Cr

Decay scheme of ^{51}Cr

^{51}Cr $t_{1/2}$: 27,7 days

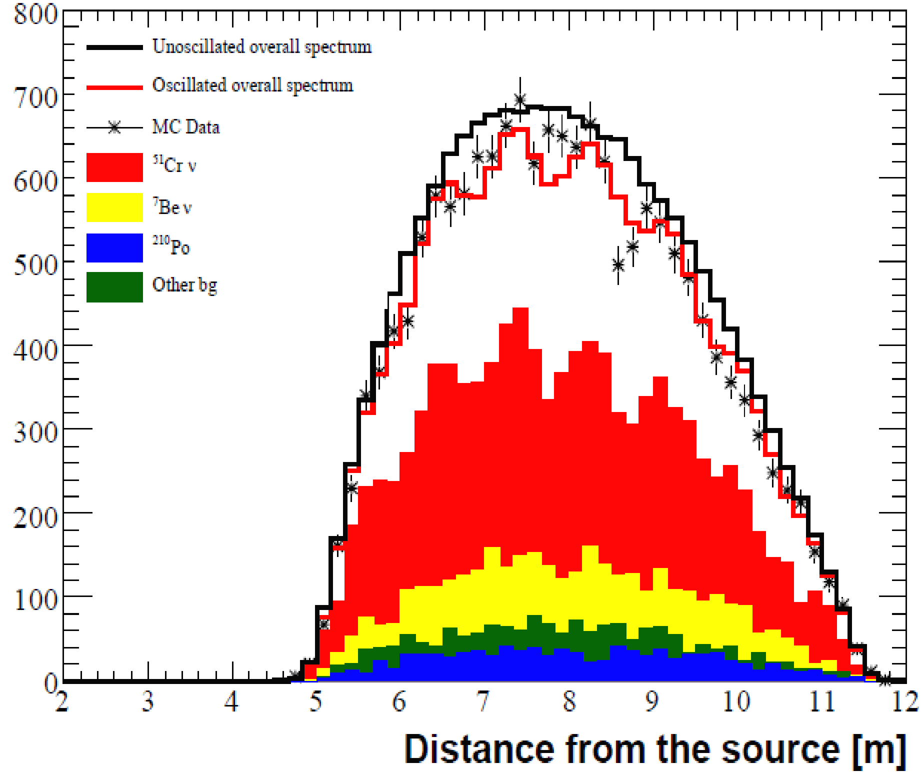
ν_1 = 427 keV (9,0%)
ν_2 = 432 keV (0,9%)



ν_3 = 747 keV (81,6%)
ν_4 = 752 keV (8,5%)

^{51}V

SoX Phase A: Possible outcome of the ^{51}Cr experiment



Production of ^{51}Cr isotope

$^{50}Cr\ (n,\gamma)^{51}Cr$

Enrichment of ^{50}Cr by gas centrifugation in form of chromium oxyfluoride $^{50}CrO_2F_2 \rightarrow {}^{50}CrO_3 \rightarrow {}^{50}Cr$ metal

Existing batch

Isotope	^{50}Cr	^{52}Cr	^{53}Cr	^{54}Cr
Natural composition	4,35	83,8	9,5	2,35
^{50}Cr - 38,6%	38,6	60,7	0,7	<0,3
^{50}Cr - 80,0%	80,0	20,0	0	0

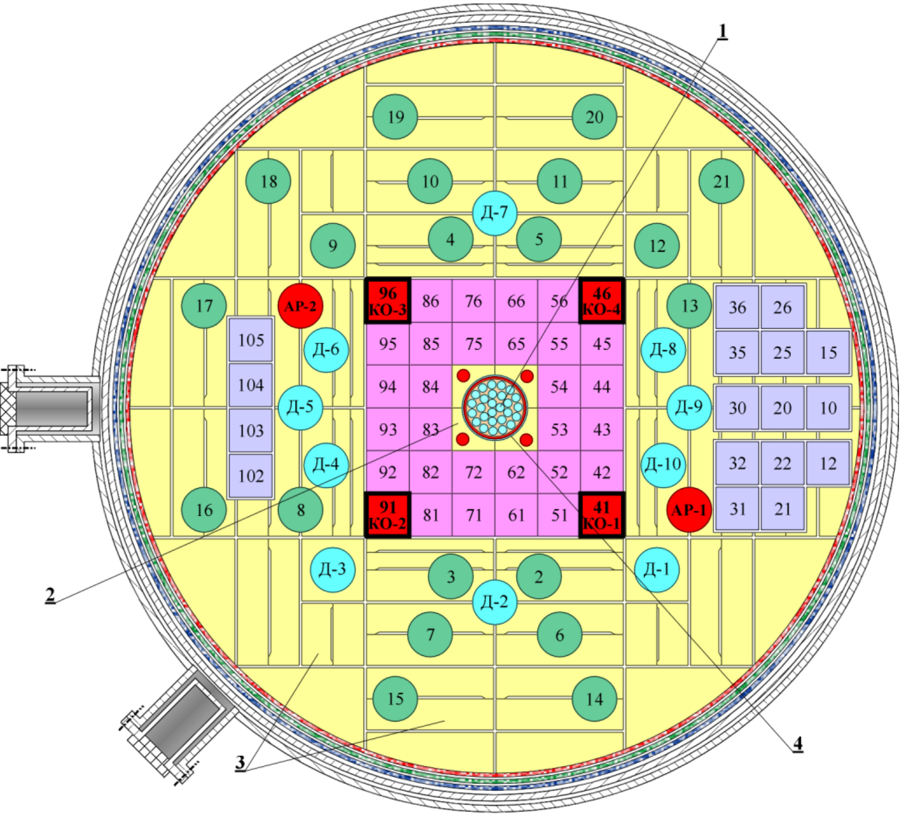
<< 1% used In GALLEX

	^{50}Cr	^{52}Cr	^{53}Cr	^{54}Cr
Thermal cross-section, barn	15,9	0,76	18,2	0,36
Resonance Integral, barn	7,8	0,48	8,9	0,18

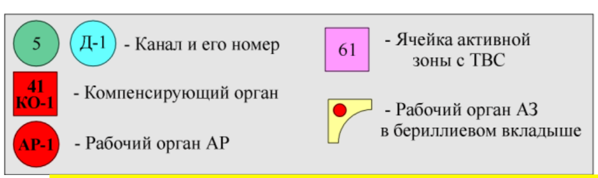
List of reactors in Russia suitable for ^{51}Cr production at MCi-scale

Reactor/ Thermal power	Neutron flux	Irradiation volume	Remarks
SM-3 100 MWt *(moderator: pressurized water/Be)*	Thermal flux $50.0 \cdot 10^{14}$ cm^{-2}s^{-1} $12.6 \cdot 10^{14}$ cm^{-2}s^{-1} $10.0 \cdot 10^{14}$ cm^{-2}s^{-1} $5.1 \cdot 10^{14}$ cm^{-2}s^{-1} $1.3 \cdot 10^{14}$ cm^{-2}s^{-1}	Neutron trap: 0.54 L; Reflector: 1.6 L x 8 = 12.8 L 1.6 L x 4 = 6.4 L 1.6 L x 10 = 16 L 1.6 L x 8 = 12.8 L	High flux research reactor (T_{cool} : input - 50^{o}C output ≤ 95 ^{o}C)
L-2 LUDMILA *(moderator: heavy water)*	Thermal flux: $1 \cdot 10^{14}$ cm^{-2}s^{-1} ⇓ $2 \cdot 10^{14}$ cm^{-2}s^{-1} (at the end of campaign)	Neutron traps: 6*1.1 L = 6.6 L Total irradiation volume: V_{tot} ~ 300 L	Heavy water reactor for isotope production (T_{cool}: input – 30^{o}C output ≤ 36 ^{o}C)
BN-600 1 500 MWt *(cooling: liquid sodium)*	Fast neutron flux: $6 \cdot 10^{15}$ cm^{-2}s^{-1} Thermal neutron flux: $2 \div 3 \cdot 10^{15}$ cm^{-2}s^{-1}	1st row of a blanket ~ 74 cells ------ Irradiation Assembly: ~ 50 cm^3 of each	Fast breeder reactor (T_{cool} ≤ 550 ^{o}C)

Option 1: SM-3 reactor with 38% ^{50}Cr chips



1 - центральный бериллиевый блок; 2 - бериллиевые вкладыши;
3 - бериллиевые блоки отражателя; 4 - центральный компенсирующий орган.

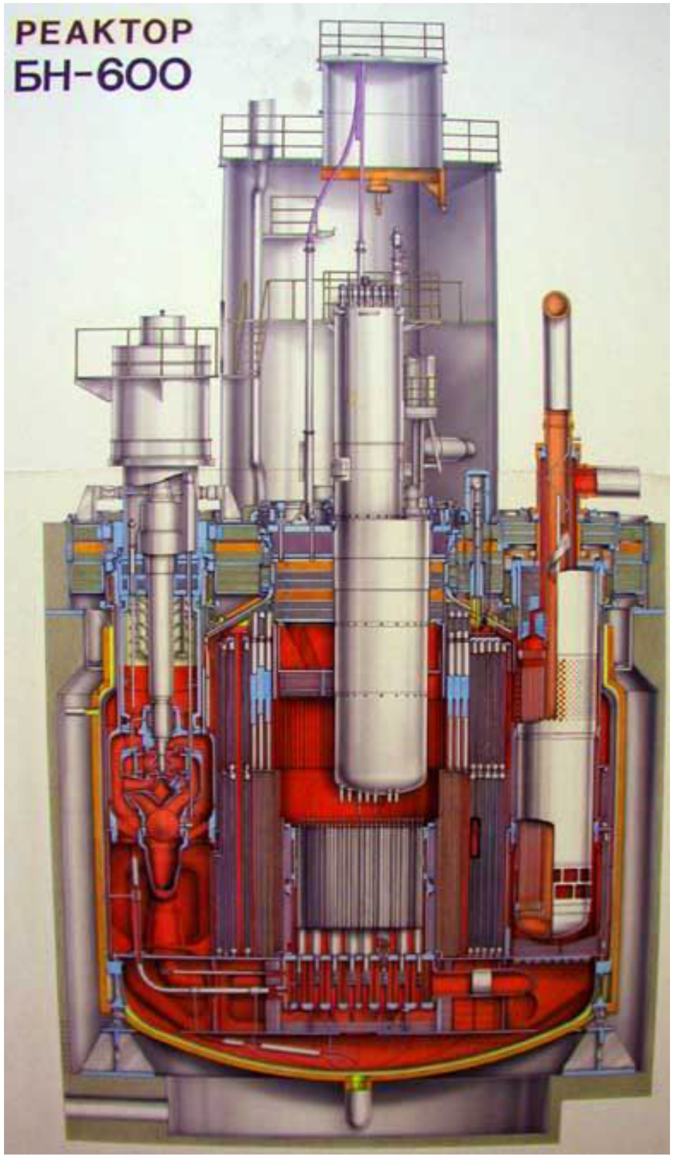


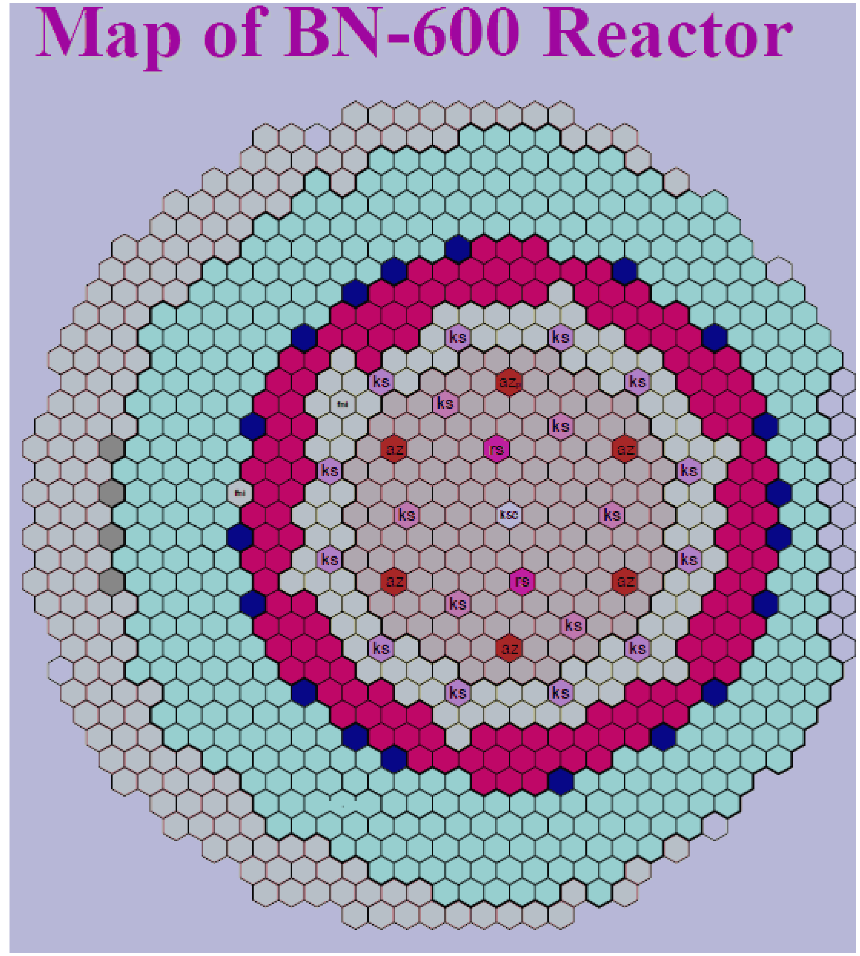
1 – Neutron water trap with 27 tubes made of Zr;
2 – Beryllium inserts of the water trap;
3 – Side reflector made of Beryllium blocks with 30 irradiation channels;
4 – Compensation rod.
Notations: ***5*** and ***Д-1*** – channel and its number;
61 – Cell with a fuel assembly;
41 КО-1 – Compensation rod;
АР-1 Automatic regulation rod;

Enrichment	**38%**
Number of reactor targets	**138**
Mass of chromium	**12 kg**
Irradiation time	**43 days**
Average specific activity (EOB)	**284 Ci/g**
Total activity (EOB)	**3.7 MCi**
Maximum total activity (36 kg of chips @ 38,6%)	**6.8 MCi**

Now SM3 is negotiating with INR RAS concerning the production of 3 MCi ^{51}Cr source for Ga experiment at Baksan (Russia)

Option 2: Reactor BN-600 of Beloyarskaya Nuclear Power Plant (Zarechny, Russia)





Note. The ^{37}Ar neutrino source with A = 0,41 MCi has been produced using BN600 for SAGE Collaboration in 2004

Option 2: ^{51}Cr production of the reactor BN-600 at 1st row of a blanket

Chromium rods	BN-600 ∅ 12 mm	
Enrichment	36%	90%
Mass of chromium, g	800	800
Average specific activity, Ci/g	484	1000
Mass of Cr to produce of 1 MCi activity, kg	2.1	1.0
Total activity in IA, MCi	0.39	0.80
Minimum quantity of Irrad.Assemblies to produce 1 MCi	3	2

Reactor campaign ~ 160 days

IA moderator ZrH_x

Absorber of thermal neutron Eu_2O_3

For an activity of 10MCi (at EOB):

Total mass of ^{50}Cr (90%) 10 kg

Number of Irradiation Assemblies 13

The reactor is devoted to the production of electricity. Extremely difficult to convince them again for production of ^{51}Cr neutrino source.

Option 3: LUDMILA reactor @ PA "MAYAK", Russia

· Moderator D_2O

Coolant D_2O

Temperature of coolant, °C (in/out) 30 °C/36 °C

Neutron flux $(1 - 2) * 10^{14}\ cm^{-2}\ s^{-1}$

· Total irradiation volume ~ up to 0.3 m^3 (~ 300 L)

· Re-loading procedure can be done at full (or a little decreased) power of the reactor.

Time to unload 35 kg of ^{50}Cr chips from 180 irradiation assemblies :

~ 10 days

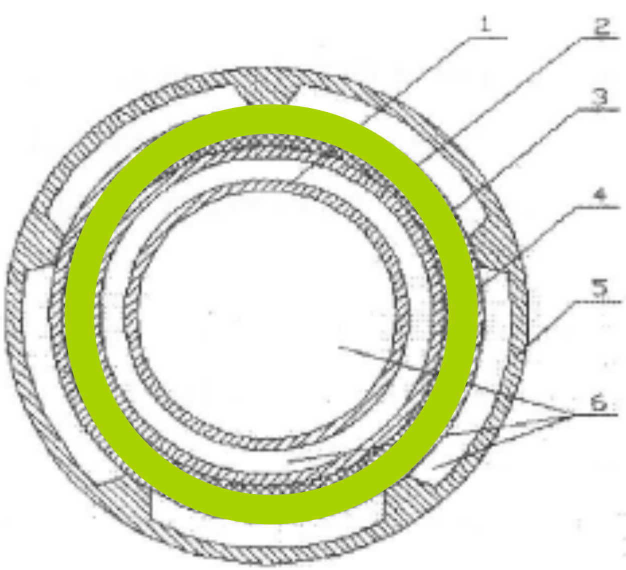
Irradiation Assembly for ^{50}Cr chips (LUDMILA)

m (^{50}Cr): 150 g

200 g

250 g

17



Zone	Thickness, cm	Inner radius, cm	Outer radius, cm	Material
1	1.5	.0000	1.50	Heavy water
2	.100	1.500	1.6	Al
3	.300	1.600	1.90	Heavy water
4	.100	1.90	2.00	Al
5	.400	2.00	2.40	Cr ships
6	.100	2.40	2.50	Al
7	.266	2.50	2.766	Heavy water
8	.174	2.766	2.940	Al
9	2.31	2.940	5.25	Heavy water

1. Inner tube - Ø32×1mm
2. Coating, - Ø40×1mm
3. Cr chips, - Ø48×4mm
4. Coating, - Ø50×1mm
5. Outer tube - Ø58.8× 1.5mm
6. Heavy water

L = 125 mm

Options of irradiation of ^{50}Cr target with enrichment of 38,6% and 80%

^{50}Cr enrich-ment, %	Mass of Cr, kg	Additional mass of Cr, kg	Number of IA*	Number of Irradiation Channels **	A, MCi, at EOB	A, MCi, 12 days
38,6	**35,5**	**-**	**178**	**18**	**9**	**6,67**
38,6	35,5	17,8	267	27	13,5	**10**
38,6+80	35,5	9,45	225	23	13,5	**10**
80	**28,3**	**-**	**142**	**15**	**13,5**	**10**

* One Irradiation Assembly: 200 gr of Cr chips
** 10 Irradiation Assemblies per Irradiation channel

Transportation of ^{51}Cr source

IAEA Limits on transport of ^{51}Cr in a type B(U) container by air:

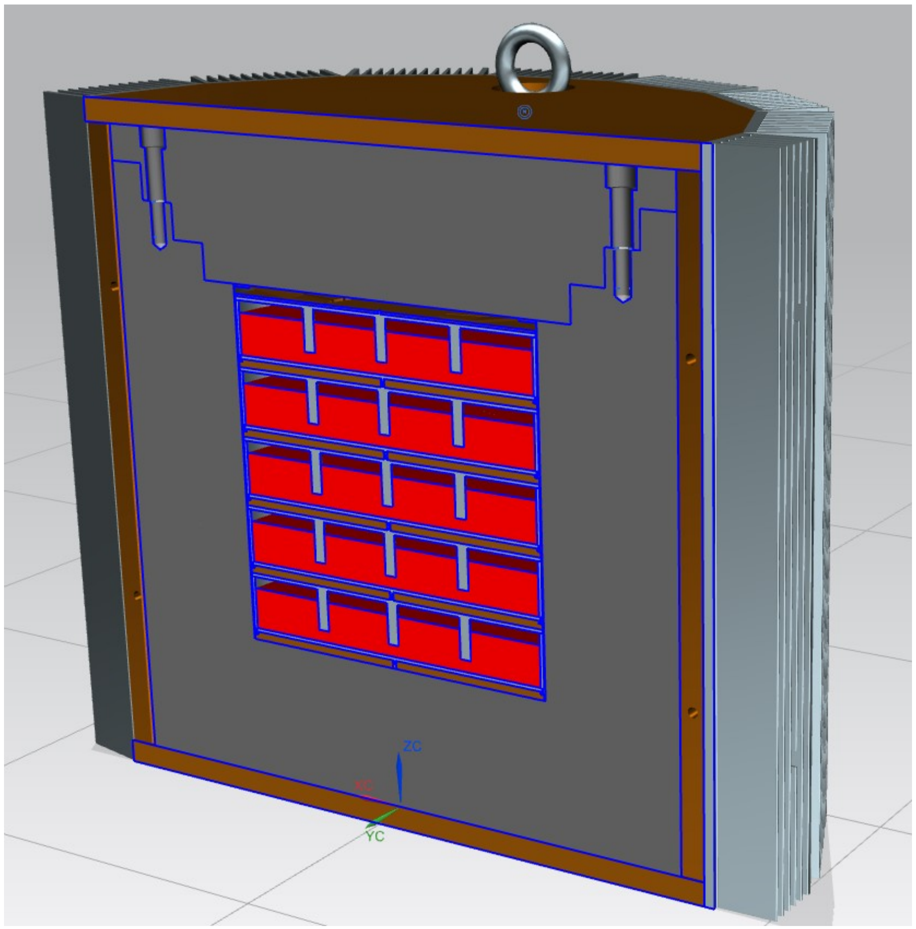
90 PBq (2,4 MCi) per individual package

4÷5 transport containers but in same plane

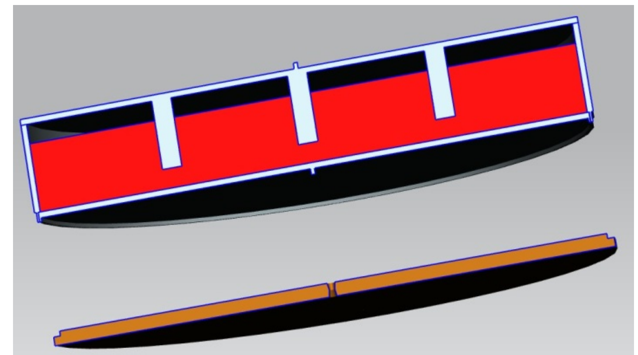
- PA Mayak – Koltsovo airport by truck (~ 140 km)
- Koltsovo – Fiumicino Roma by plane
- Fiumicino – to Casaccia Research Center (ENEA) by truck (**to combine 4÷5 sources into single one**)
- Casaccia – LNGS by truck

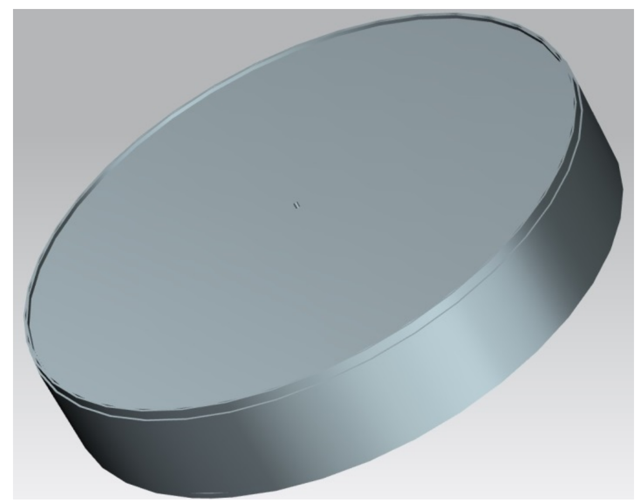
Possible design of ^{51}Cr source consisted of 4 – 5 individual sources with activity of 2,4 MCi each

Dimensions
D50 mm x 530 mm (H)



Individual source
D290 mm x 50 mm





Recipe for an Antineutrino generator $^{144}Ce/^{144}Pr$

CeLAND experiment :

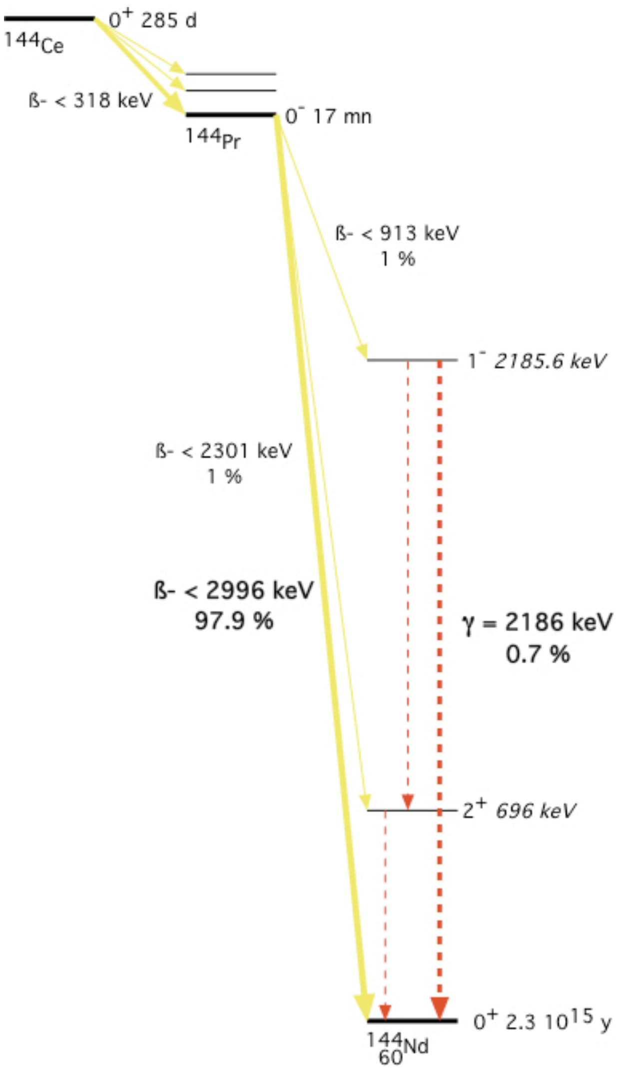
Phase I (2015)	**75 -100 kCi in water tank or in Xe room**
Phase II (2016/2017) ?	**50 kCi inside detector**

SoX experiment:

Phase B (2015 ?)	**75 kCi in water tank**
Phase C (2016/2017)	**50 kCi inside detector**

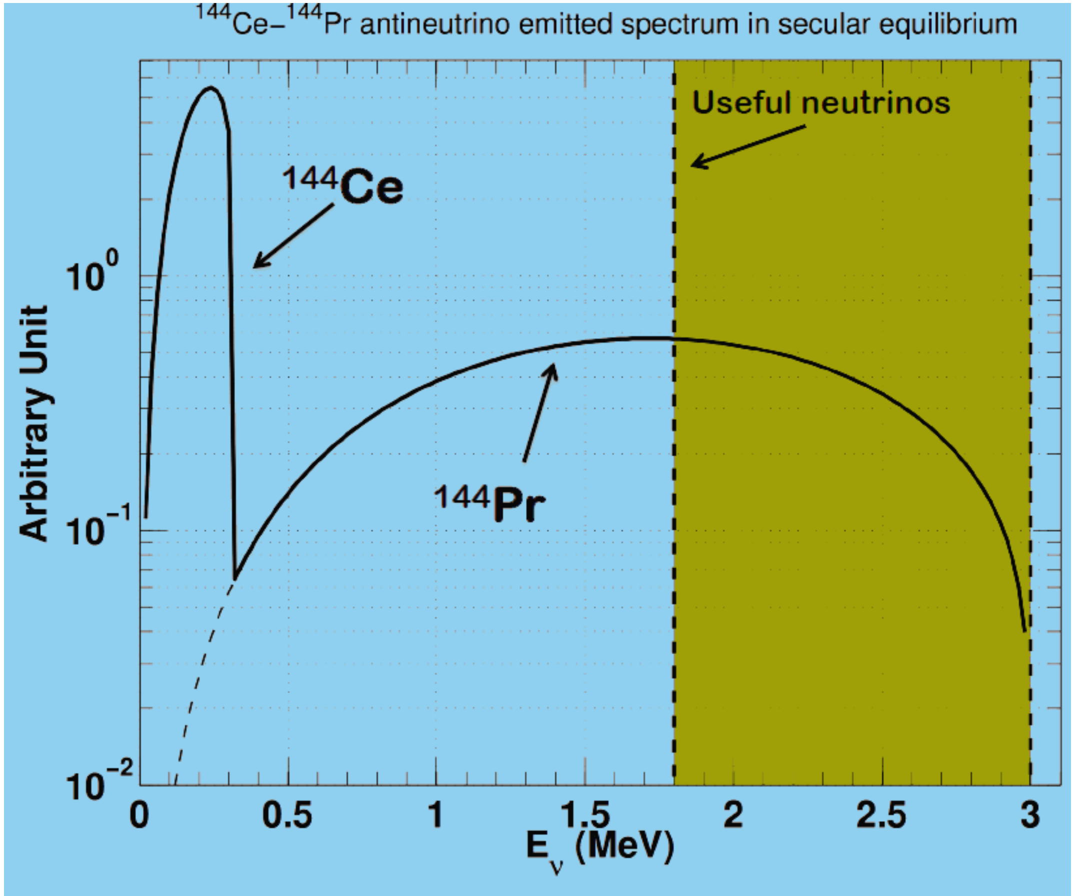
Competition or/and mutual efforts?

$^{144}Ce/^{144}Pr$ antineutrino spectrum



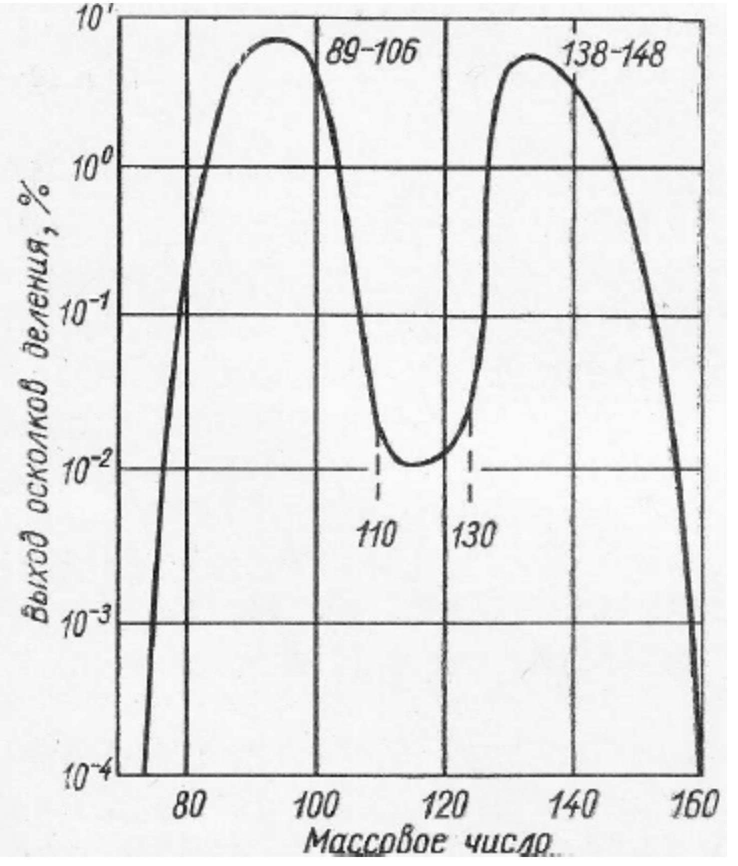
Detection via IBD

$\overline{\nu} + p \rightarrow e^{+} + n \; (E_{thr} = 1{,}8 \text{ MeV})$



Cerium : an abundant fission product

Yield, %



A

1st maximum: Br, Kr, Rb, **Sr** (***5,8%***), Y, Zr, Nb, Mo, Tc, **Ru** (***0,4%)*** = 38,5%

2d maximum: Te, I, Xe, Cs, Ba, **Ce** (***5,5%***), Pr, Nd, Sb = 32,8%

In 1 ton of SNF (spent nuclear fuel)

*reactor VVR-440, burning of 40 GW*days/t*

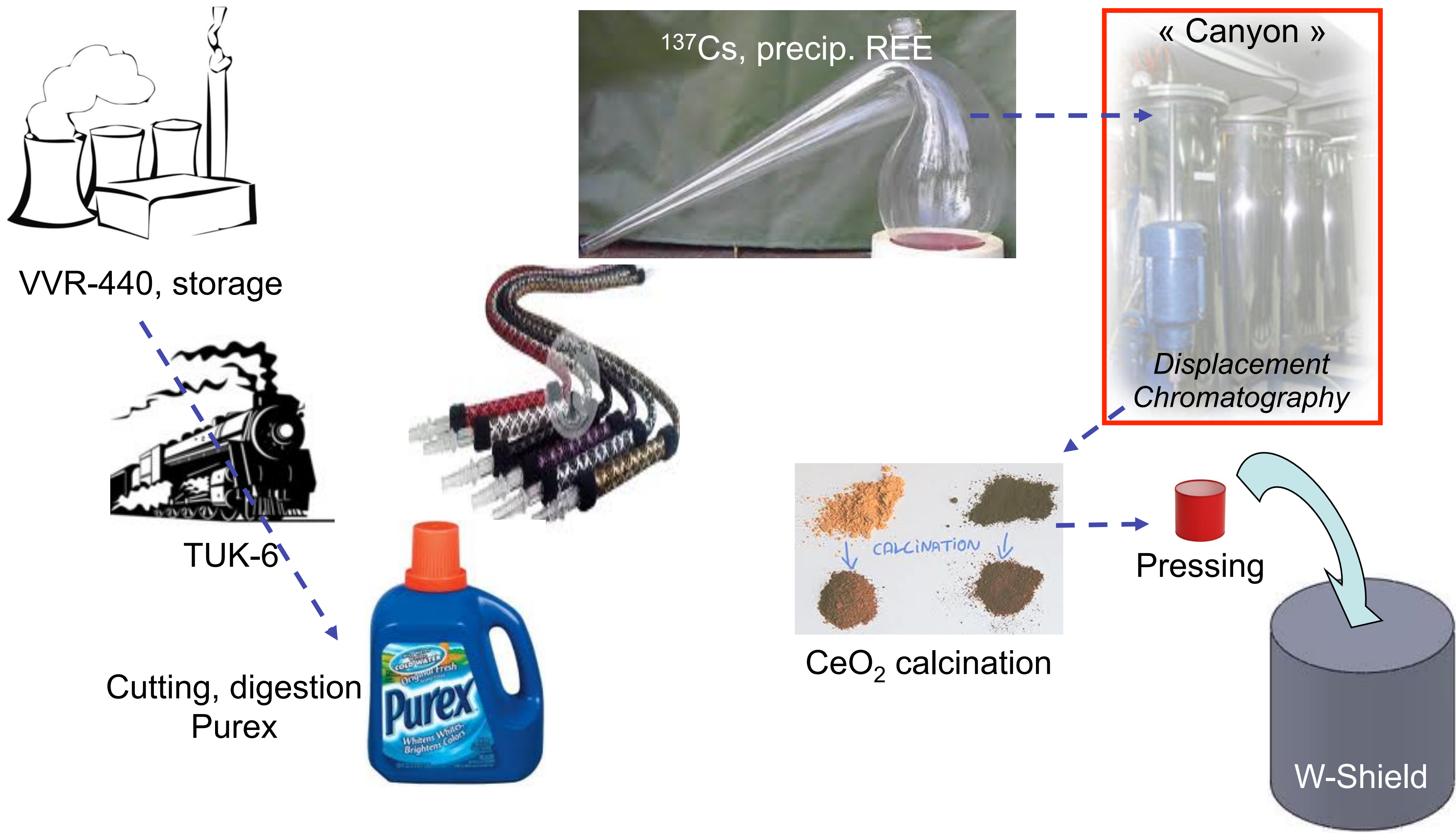
- **Pu239** **10 kg**
- **Np237** **0,7 kg**
- **TPE (Am, Cm)** **0,6 kg**
-
- **Fission product** **44 kg,**

including Rare Earth El. + TPE ~ 13,2 kg (Ce ~ 22%)

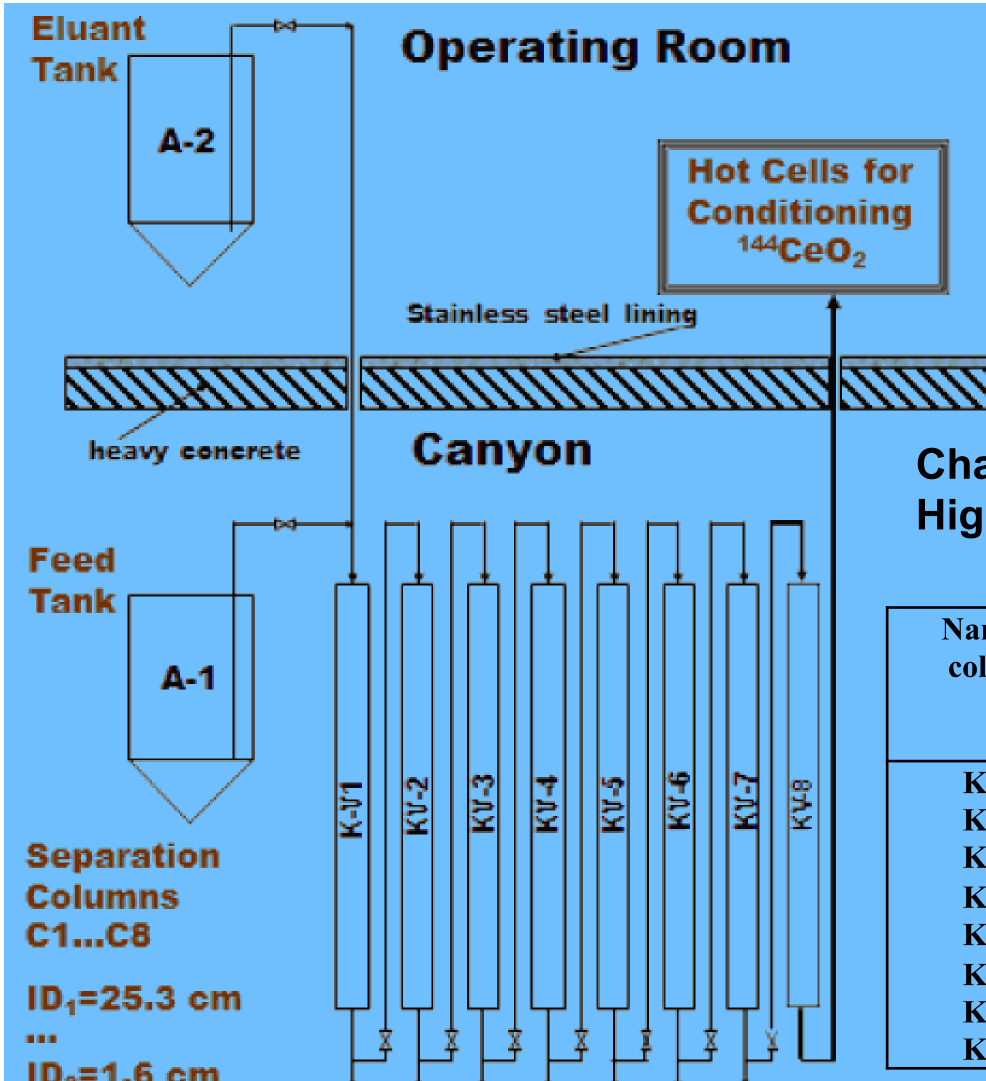
^{144}Ce/(sum of Ce isotopes) = 1:200 ÷1:110

depends strongly on history of given fuel element during reactor campaign

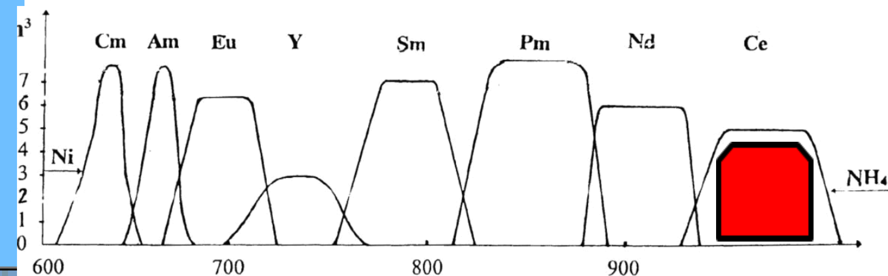
Scheme of production



The chromatographic facility for separation of REE and TPE at PA Mayak



Output Separation Curve of REE and TRU Group



Characteristics of chromatographic columns
Hight = 2,5 m (columns №№ 1 ÷ 6)
= 2,0 m (columns №№ 7 и 8)

Name of column	Cross-section of column, cm^2	Total volume, L	Filtration velocity under consumption of 4 $mL/min*cm^2$, L/hour
KV-1	502,9	125,7	120,7
KV-2	323,8	81,0	77,7
KV-3	169,8	42,4	40,8
KV-4	75,5	18,9	18,1
KV-5	34,2	8,6	8,2
KV-6	17,3	4,4	4,2
KV-7	5,1	1,0	1,2
KV-8	2,0	0,4	0,5

Chromatographic facility for ^{147}Pm recovery and purification at PA MAYAK (Ozersk)

Productivity = 10 kg of Rare Earth Elements per cycle

Duration of one cycle ~ 15 days

Content of Ce element ~ 22%

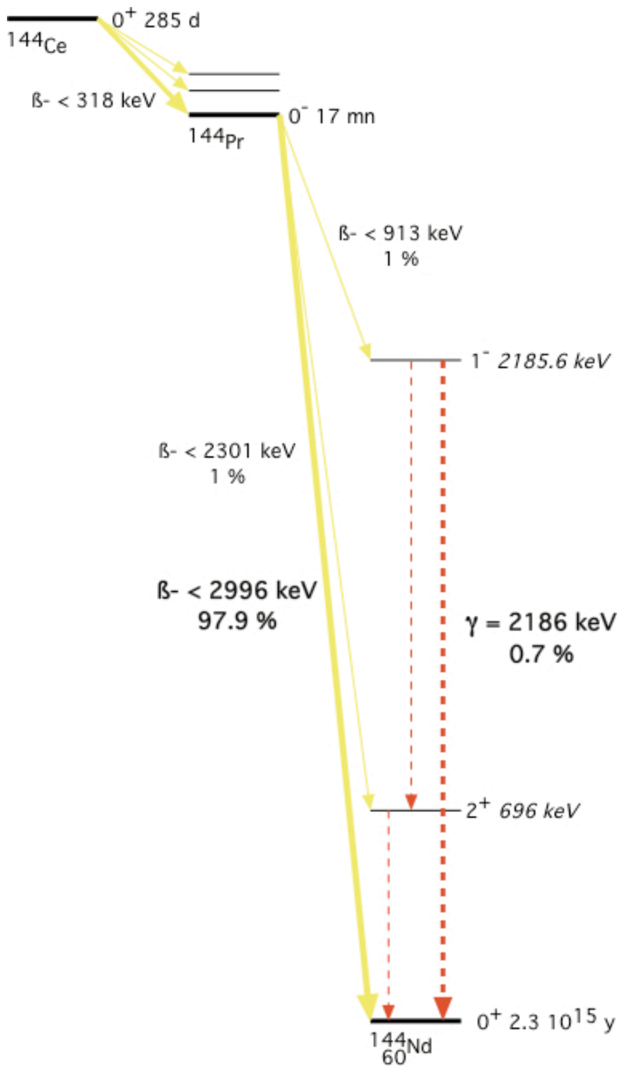
Purity of Pm (Ce) in others REE (γ-emitters)

$\leq 10^{-8}$

Purity of Pm (Ce) in any others TPE (neutrons emitters)

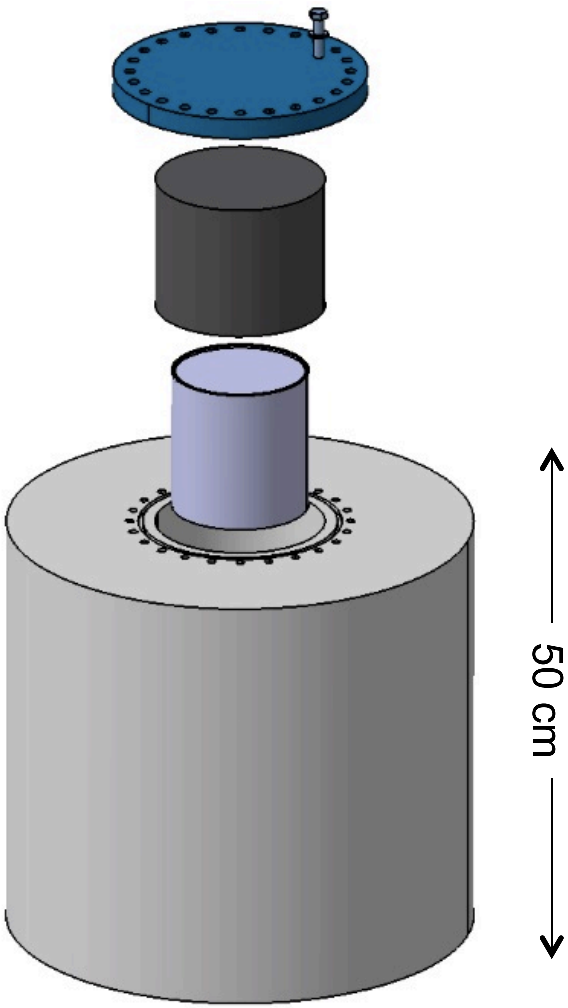
$\leq 10^{-8}$

^{144}Ce-^{144}Pr



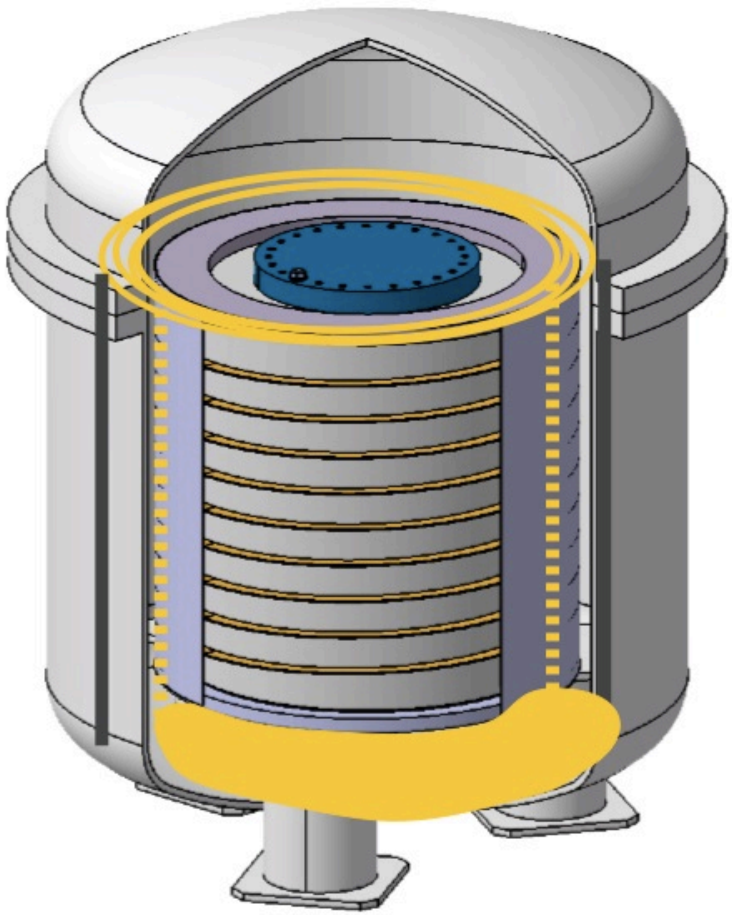
- Cerium, Praseodymium, Neodymium
- mean life : 411.04 ± 0.07 d ; -5% in 3 weeks
- 75 kCi (2.77 PBq)
  - 23.56 g ^{144}Ce
  - ≈ 3 - 8 kg of CeO_2 history of SNF
- density : 4.0 ± 1.0 g/cm^3
- 75 kCi => 599 W
  - <power>1y = 352 W
  - 7.991 ± 0.044 W/kCi ie 0.56 %
- after 1.5 year 158 W (*210 W*)

Characteristics of the source



- 75 kCi = 2.77 PBq (up to 100 kCi)
  - ≈ 8 kg of CeO_2 (ρ= 4 g/cm^3)
  - 2.0 liters
- Inner double sealed stainless steel cylinders
- Cylindrical tungsten shield
  - Companies in Europe, China, Russia
  - Densimet® 185 or similar product
    - 18.5 g/cm^3 ; 97%W +Fe,Ni
  - water tight upper cork
  - 47 cm x 47 cm
  - 16 cm width minimum
  - 1.5 ton and ext. surface ≈ 1 m^2
- Dose rate (scaled estim. by SPR)
  - 1.5 mSv/h at contact
  - 50 μSv/h at 1m from shielding
- Estimated temperature at surface
  - 599 W ; external temp. 38 °C
  - ≈ 100 °C in open air

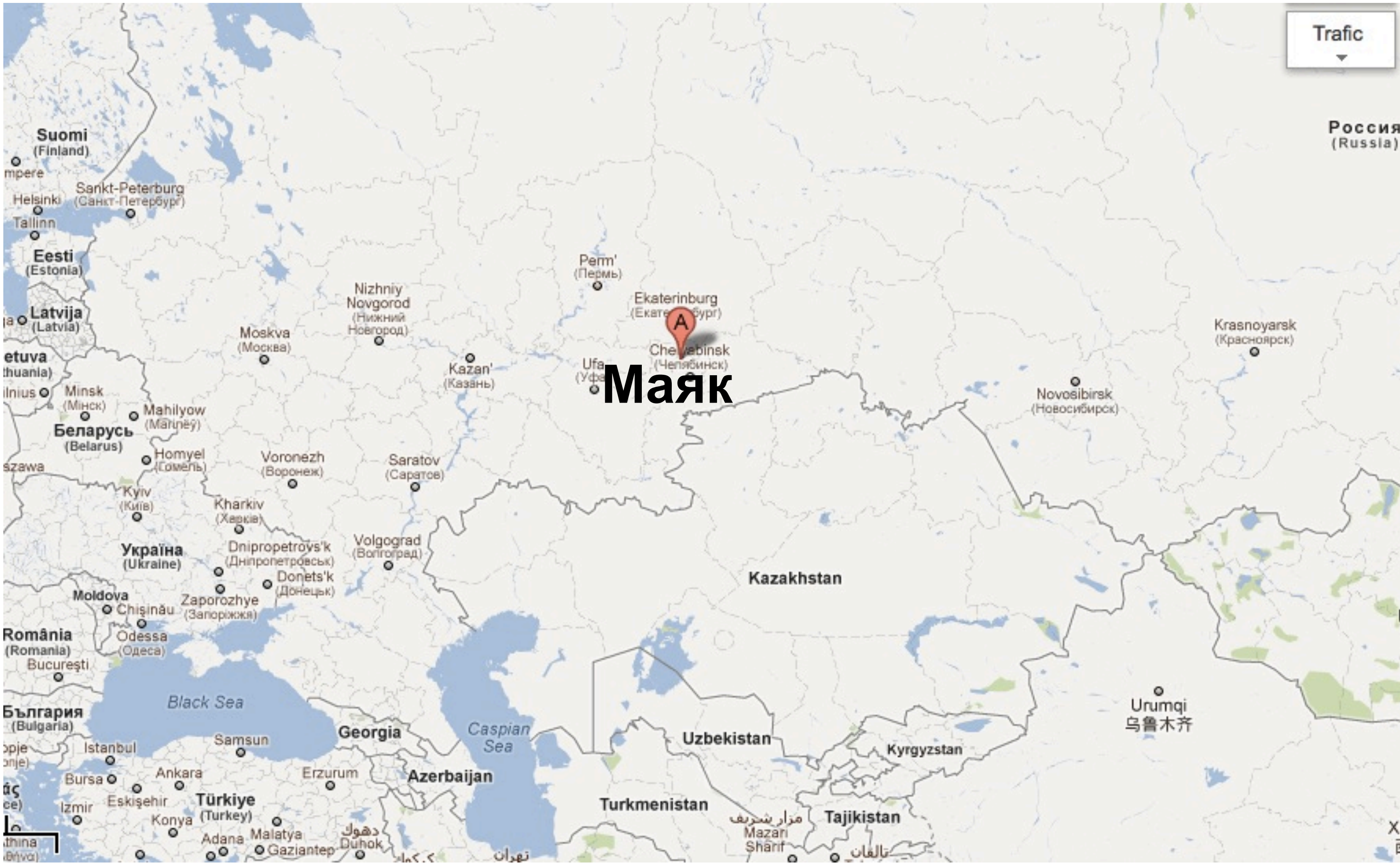
Measure the activity by « real » calorimetry



Water cooling (600 W)
- $\Delta T = 40$ K
- Fixed cold point
- Flow rate ≈ 12 lit/hr

Very good insulation toward exterior :
- Suspension by kevlar ropes : minimize conduction
- Vacuum (10^{-6} bar) in the vessel : minimize convection
- Multilayer wrapping : minimize radiation

In the Oural mountains



Possible routes for transport

- Severe constraints based on regulation issued by IAEA
  - nothing impossible, but long, bureaucratic and costly
  - by air limit for each radioisotope : 16.2 kCi for ^{144}Ce, (2.4 MCi for ^{51}Cr)
  - by boat : only limited number of harbours agreed for radioactive materials

No satisfactory solutions so far !

CONCLUSION 1: ^{51}Cr source & Borexino

- **With existing batch of ^{50}Cr (35,5 kg @ 38,6%) nobody can produce 10 MCi *on site ie* 12 days after EOB**
- **LUDMILA reactor: 9 MCi at the EOB**
- **Negotiations with the Administration of SM-3 and BN-600 reactors are extremely difficult**
- **Air transportation : IAEA rules limits ^{51}Cr activity at 2,4 MCi per individual B(U) package, but several package in same plane possible**
- **Need job at Casaccia Research Center (ENEA) to combine 4÷5 sources into single one for further shipment to LNGS.**

CONCLUSION 2: ^{144}Ce/^{144}Pr source for CeLAND and SoX

- **^{144}Ce/^{144}Pr source with activity in the range of 50 kCi till 100 kCi can be produced at PA "Mayak" (Ozersk, Russia).**
  PA "Mayak" reprocesses up to 300 tons of SNF per year (mainly from PWR VVER440).
- **Strongly recommended to use SNF after cooling time at Kola NNP not longer than 3 years.**
- **Productivity of existing chromatographic facility is 10 kg of REE +TPE (or about 2 kg of Ce) per cycle**
- **Ratio ^{144}Ce/Ce element is in the range of 1:200 to 1:110.**
  Mass of Ce (element) = 3,1 ÷ 8,0 kg function of history of SNF used
- **Transportation issue:**
  - **IAEA air transport limits ^{144}Cr/^{144}Pr 600 TBq (16,2 kCi)**
  - **Longer lifetime : surface transport (train, boat, trucks) possible. No constraint on activity**

Thank you for your attention !

Backup slides

List of reactor in USA and Europe suitable for Cr51 production at MCi-scale

Reactor/ Thermal power	Neutron flux	Irradiation volume	Remarks
HFR 45 MWt *(moderator: pressurized water/Be)* Netherland	Thermal flux $5.6 \cdot 10^{14}$ cm^{-2}s^{-1}	In core: 19 channels In reflector: 12 channels	High flux research reactor 12 campaigns/year
HFIR 85 MWt Oak Ridge *(moderator: pressurized water/Be)* USA	Thermal flux: $2{,}5 \cdot 10^{15}$ cm^{-2}s^{-1}	Neutron traps: D12,7 x 61 cm	Heavy water reactor for isotope production (T_{cool}: 49 /69 ^{o}C)
BR-2 100 MWt *(moderator: pressurized water/Be)* Belgium	Thermal neutron flux: 10^{15} cm^{-2}s^{-1} $2 \div 3{,}5 \cdot 10^{14}$ cm^{-2}s^{-1}	7 channels in Be plug: D 200 mm D 84 mm D 50&33 mm	High flux material testing reactor

LUDMILA reactor L2 (PA MAYAK)

The large irradiation volume in active zone gives the possibility to place in reactor core ***up to a few tens of kg*** of start material and even to place 36 kg of chromium chips in the form of thin layers to avoid the effect of 50Cr self-shielding.

The large integral flux of excess thermal neutrons makes it possible to create a ***51Cr ANS with activity *8MCi (and 10 MCi due to special neutron «traps»***); ***since Dec 2011: + 20% and more (new campaign)!***

The convenient thermal conditions exclude the possibility of ***sintering effect for chromium chips coagulating*** during irradiation.

The construction of Irradiation Assemblies allows re-loading of channels at the full or slightly decreased power level. This gives great flexibility in unloading irradiated material in accordance with a given channel's productivity and with the lifetime of isotope, thereby minimizing the loss due to its decay.

The reactor L-2 has negative feedback reactivity effects (power and temperature) ensuring reliable control and a high level of safety.

Self-shielding coefficient of the thermal neutron flux into Irradiation Assembly with Cr

Enrichment on ^{50}Cr, %	Mass of ^{enr}Cr, g		
	150	**200**	250
4,35	0,937	**0,922**	0,908
38,6	0,885	**0,859**	0,836
80,0	0,819	**0,780**	0,746

Pressurized Water Reactor VVER-440 (Russia)

- **Number of reactors (VVER-440) 6**
- **Production rate of SNF per year 87 t**
- **Number of Fuel Elements with SNF ~ 700**

- “Closed nuclear uranium cycle” of SNF is implemented in Russia:

  Temporary storage at APS (at special storages) during 3 – 5 years, and then they are dispatched/transported to the radiochemical plant RT-1 (PA “Mayak”, Ozersk).

Note. The RT-1 radiochemical plant is only facility in the world where each individual chemical element from SNF can be recovered and purified.

Typical process to recovery of individual element which belongs to Rare Earths group of SNF

~ 2 - 3 t of Spent Nuclear Fuel

⇓

Purex process
(removal of *U,* Np and *Pu*) ⇒ *production of nuclear fuel*

⇓

Evaporation of HNO_3 ⇒ *vitrification of nuclear waste*

⇓

Purification of REE and TPE concentrate of ^{137}Cs, ^{90}Sr, Ni, Cr, ..impurities by oxalate precipitation

⇓

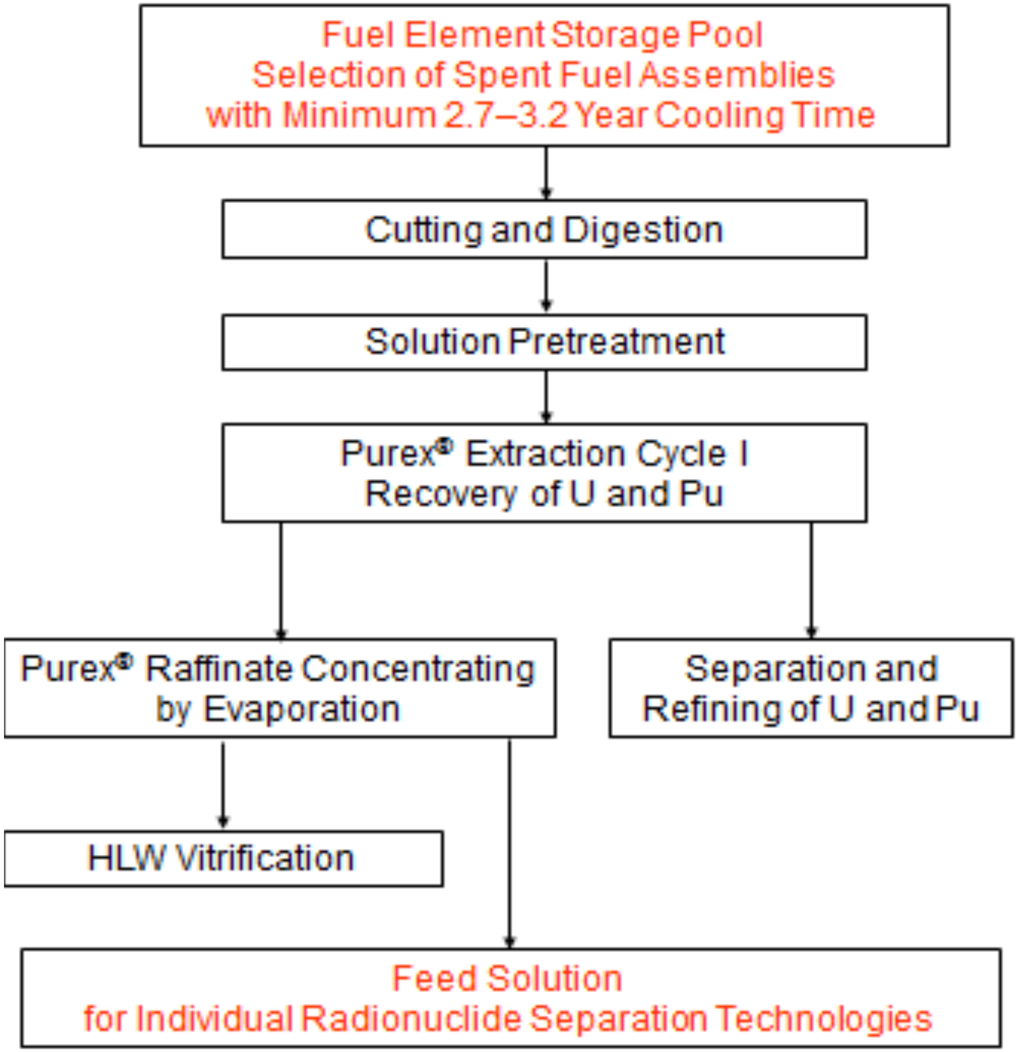
Chromatography purification
(recovery and purification of Cerium fraction)

⇓

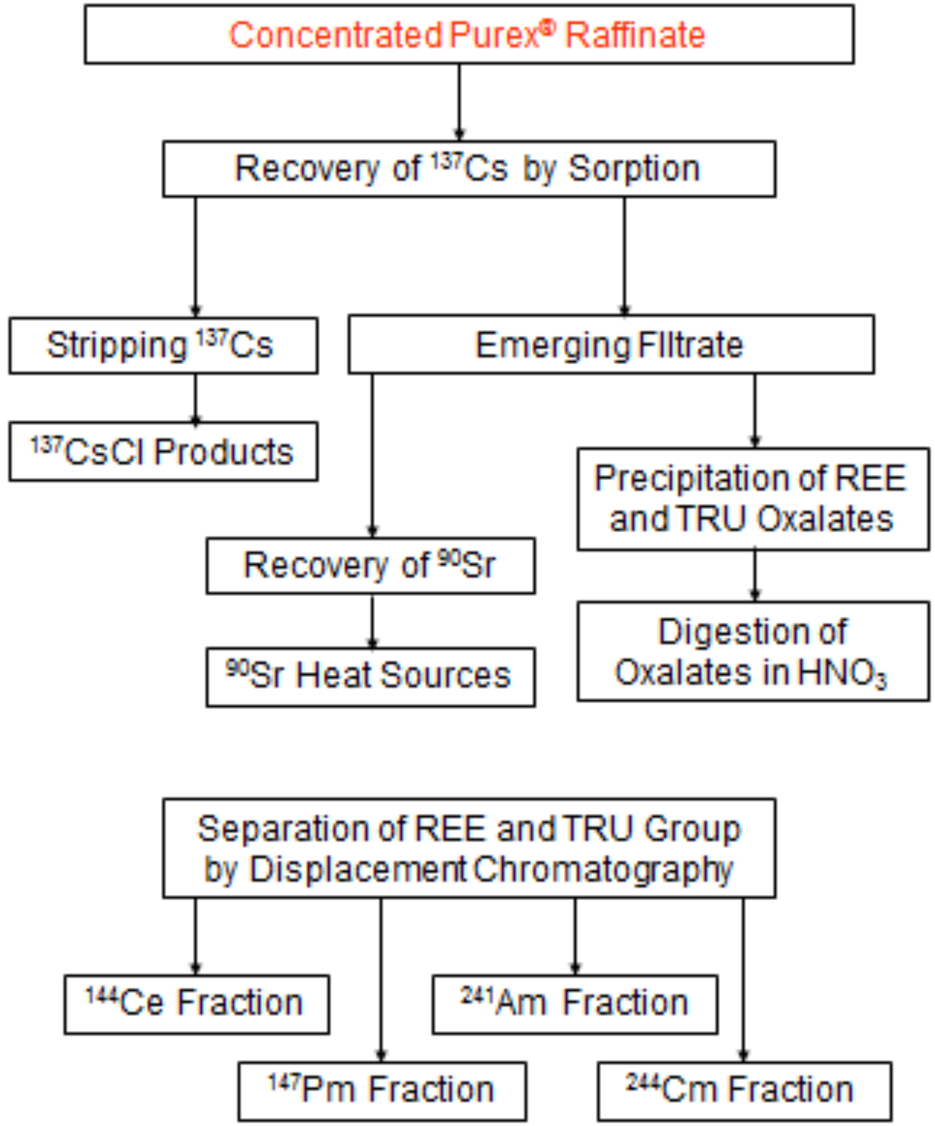
Ce-oxalate precipitation

⇓

CeO_2
(t ~ 850 ^{o}C)



Phase I: Selected Spent Fuel Processing to Obtain the Feed Required



Phase II: Individual Radionuclide Production – Separation and Refining

Solution of ^{144}Ce

↓

Precipitation of $^{144}Ce_2(C_2O_4)_3$
by Oxalic Acid

↓

$^{144}Ce_2(C_2O_4)_3$ Calcining $\rightarrow$ $^{144}CeO_2$

↓

$^{144}CeO_2$ Encapsulating

↓

Fabrication of ^{144}Ce Ionizing Source
D=120 mm, H=380-440 mm

↓

Transportation of ^{144}Ce Ionizing
Source in the Shielding Container to
the Customer

Phase III: Individual Radionuclide Production – Ionizing Source Fabrication and Handling